Two-Dimensionality of Magnetic Excitations on the Trellis Lattice: $(La,Sr,Ca)_{14}Cu_{24}O_{41}$ and $SrCu_2O_3$

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We explore the properties of magnetic excitations on the trellis lattice which is relevant for the so-called telephone-number compounds $A_{14}Cu_{24}O_{41}$ and the system $SrCu_2O_3$. The trellis lattice consists of two-leg ladders which are coupled in a strongly frustrated fashion. We use the effective model obtained for a single two-leg spin ladder to calculate the two-dimensional one-triplon dispersion and the corresponding one-triplon contribution to the dynamical structure factor. Special attention is laid on signatures of the frustrating inter-ladder magnetic exchange. A detailed suggestion is made for an experimental detection of this exchange in inelastic neutron scattering experiments.

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Low dimensional quantum antiferromagnets display a variety of fascinating properties. The reduced dimensionality can either be realized by strongly anisotropic magnetic exchanges or by a strongly frustrating topology of the system. The so-called telephone-number compounds $A_{14}Cu_{24}O_{41}$ and the system $SrCu_2O_3$ are often discussed as realizations of idealized quasi one-dimensional two-leg ladders[1,2,3,4,5,6,7]. These one-dimensional structures build a two-dimensional trellis lattice (see Fig. 1), i.e. the coupling between the ladders is strongly frustrated. But the detailed influence of such a coupling and its quantitative strength are currently unknown.

Quantum chemistry calculations have revealed a sizable value for the magnetic exchange between the one-dimensional ladder structures for the system $SrCu_2O_3$[8]. It is therefore an important issue to clarify the impact of the two-dimensionality of these systems on their magnetic properties. Theoretically, this question has been treated by mean-field theory using the bond-operator approach which is reliable when the rung-coupling $J_\perp$ dominates[9]. Exact diagonalization studies were performed on small clusters which provide only a limited number of points in momentum space[10]. In this work we approach this issue by studying the effects of the inter-ladder coupling $J_{\rm int}$ on the elementary excitations of the single ladder and its dynamics. We concentrate on the paramagnetic case which is the experimentally relevant one. We give a detailed description for a possible experimental detection and determination of such a coupling by inelastic neutron scattering.

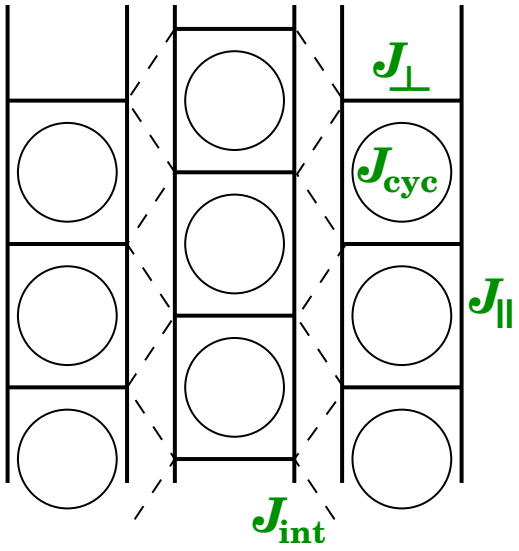

FIG. 1: Heisenberg exchange couplings ($J_\perp, J_\parallel, J_{\rm int}$) and the four-spin exchange coupling $J_{\rm cyc}$ (circles) considered in the Hamiltonian (1). The rung coupling (horizontal solid lines) is $J_\perp$, the one along the legs of the ladders (vertical solid lines) is denoted by $J_\parallel$ and the coupling between adjacent ladders (dashed lines) is denoted by $J_{\rm int}$.

The couplings considered are shown in Fig. 1. They refer

to the Hamiltonian

$$H = H_0 + H_{\rm cyc} + H_{\rm int} \tag{1a}$$

$$H_0 = J_\perp \sum_{i,j} \mathbf{S}_{i,j;\mathrm{L}} \mathbf{S}_{i,j;\mathrm{R}} + J_\parallel \sum_{i,j;\tau\in\{\mathrm{L.R}\}} \mathbf{S}_{i,j;\tau} \mathbf{S}_{i+1,j+1;\tau} \tag{1b}$$

$$H_{\rm cyc} = J_{\rm cyc} \sum_{i,j} \Big[(\mathbf{S}_{i,j;\mathrm{L}} \mathbf{S}_{i+1,j+1;\mathrm{L}})(\mathbf{S}_{i,j;\mathrm{R}} \mathbf{S}_{i+1,j+1;\mathrm{R}}) + (\mathbf{S}_{i,j;\mathrm{L}} \mathbf{S}_{i,j;\mathrm{R}})(\mathbf{S}_{i+1,j+1;\mathrm{L}} \mathbf{S}_{i+1,j+1;\mathrm{R}}) - (\mathbf{S}_{i,j;\mathrm{L}} \mathbf{S}_{i+1,j+1;\mathrm{R}})(\mathbf{S}_{i,j;\mathrm{R}} \mathbf{S}_{i+1,j+1;\mathrm{L}}) \Big] \tag{1c}$$

$$H_{\rm int} = J_{\rm int} \sum_{i,j} \mathbf{S}_{i,j;\mathrm{R}} \Big[\mathbf{S}_{i+1,j;\mathrm{L}} + \mathbf{S}_{i,j-1;\mathrm{L}} \Big] \tag{1d}$$

where the pair $i, j \in \mathbb{Z}$ denotes the rungs, not the sites, counted along the unit vectors e_1 and e_2 shown in Fig. 2. The subscripts L and R stand for the left and right spin, respectively, on the particular rung.

In order to explain the influence of various changes of parameters we give below the formulae for the two-dimensional dispersion $\omega_{k,q}$ on a trellis lattice and for the dynamic structure factor $S_{k,q}(\omega)$. The approach starts from the effective model that we have derived previously for a single ladder[11,12]. It is given in terms of triplons, the elementary excitations of the spin ladder. In the following, we give a summarized description how we derived the effective model for the single spin ladder. Technical details are given in Refs. 13,14.

We apply a particle-conserving perturbative continuous unitary transformation (CUT) which uses the states on isolated rungs as reference. The CUT maps the Hamiltonian of the single ladder to an effective Hamiltonian which conserves the number of triplons. In order to calculate spectral properties and to treat the inter-ladder coupling $H_{\rm int}$ the relevant observables have to be transformed by the same CUT. Here we use the perturbative realization of CUT which gives series expansions for the effective Hamiltonian and the effective observables in terms of the expansion parameters $x := J_\parallel/J_\perp$ and $x_{\rm cyc} := J_{\rm cyc}/J_\perp$ in the thermodynamic limit. The obtained series are extrapolated giving reliable results in a broad range of the parameter space[15,16].

For the present purpose, we need to know ω_k^0, the single-triplon dispersion along the ladder and the spectral weight a_k^2 of the single triplon when it is excited by S^z or another spin component. This spectral weight is known as function of the wave vector k. The coupling between the ladders is treated on the mean-field level starting from the ladders, not the isolated rung dimers[9], as unperturbed system[17,18]. We stress that the hardcore property of the triplons is taken into account along the ladders but not perpendicular to them.

The rungs of the trellis lattice form a Bravais lattice so that there is exactly one mode of given magnetic quantum number S^z per point in the first Brillouin zone. But the lattice has a skewed unit cell due to the fact that adjacent ladders are shifted with respect to each other by half the distance c between two rungs in one ladder (see Fig. 2). Conventionally, one would express the momentum in units of the reciprocal

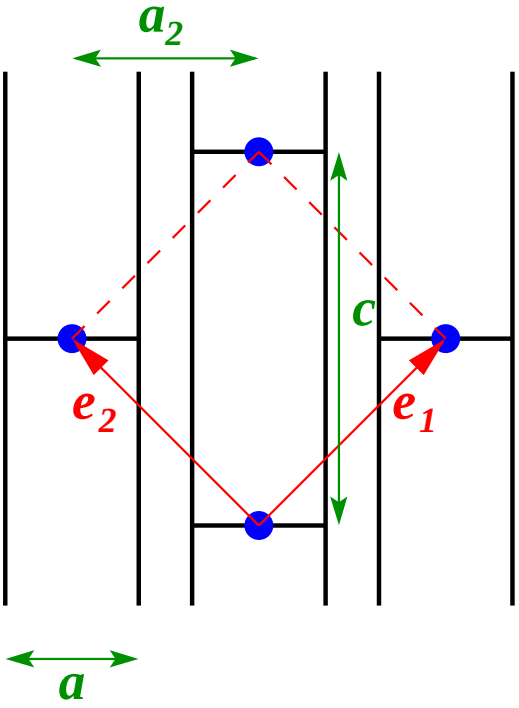

FIG. 2: Unit cell of a trellis lattice spanned by the unit vectors e_1 and e_2. The distance between two adjacent rungs on the same ladder is c. The distance between the legs of a ladder is a, the distance perpendicular to the ladders between the centers (blue dots) of two rungs on adjacent ladders is a_2.

basis vectors e_1^* and e_2^*. But in this system it is more convenient to describe the momentum by its component along the ladders (k, measured in units of $2\pi/c$) and by its component perpendicular to the ladder (q, measured in units of $2\pi/(2a_2)$, cf. Fig. 2). Note that as a consequence the dispersion $\omega(k,q)$ does not have the periodicities one would naively expect. The periodicity in k and q separately is 2, not unity. But shifting $k \to k+1$ and $q \to q+1$ together reproduces the spectrum as it has to be.[22]

We use the same approach as in Refs. 17,18 to treat the coupling $H_{\rm int}$ between the ladders. The observables $S_i^{x,y,z,\mathrm{R}}$ are transformed according to

$$S_{i,\mathrm{eff}}^{x,y,z,\mathrm{R}} := U^\dagger S_i^{x,y,z,\mathrm{R}} U = \sum_\delta a_\delta \, (t_{i+\delta}^{x,y,z,\dagger} + t_{i+\delta}^{x,y,z}) + \ldots \tag{2}$$

where the dots stand for normal-ordered quadratic and higher terms in the bosonic operators. In the following we neglect these terms corresponding to multi-triplon contributions. Note that the symmetry $S_{i,\mathrm{eff}}^{\alpha,\mathrm{L}} = -S_{i,\mathrm{eff}}^{\alpha,\mathrm{R}}$ holds on this level of approximation. The Fourier transform squared of a_δ yields the one-triplon spectral weight a_k^2. In this approximation, the total Hamiltonian of the trellis lattice is quadratic in terms of the operators $t^{x,y,z,\dagger}$ and $t^{x,y,z}$. Neglecting the hardcore property allows us to diagonalize the Hamiltonian by a Bogoliubov transformation which is justified as long as the coupling $J_{\rm int}$ is small.

The following two-dimensional dispersion $\omega_{k,q}$ is obtained

$$\omega_{k,q} = \omega_k^0 \sqrt{1 - \frac{8J_{\rm int}}{\omega_k^0} a_k^2 \cos(\pi q)\cos(\pi k)} \ . \qquad (3)$$

The two-dimensional dynamic structure factor reads

$$S_{k,q}(\omega) \ = \ a_{k,q}^2 \, \delta(\omega - \omega_{k,q}) \qquad (4a)$$

$$a_{k,q}^2 \ = \ 2\sin^2\left(\pi q \frac{a}{2a_2}\right) a_k^2 \frac{\omega_k^0}{\omega_{k,q}} \ . \qquad (4b)$$

The sine factor stems from the interference of the excitation processes from the left and from the right spin in each rung. Recall that a single triplon has an odd parity on the isolated ladder with respect to reflections about the centerline[11,12].

The data for an isolated ladder is shown in Fig. 3 for the one-triplon dispersion of a single ladder and in Fig. 4 for the one-triplon weight. The couplings and energies are given in units of the rung coupling $J_\perp$, that is $x := J_\parallel/J_\perp$, $x_{\rm cyc} := J_{\rm cyc}/J_\perp$. This constitutes the input data for the two-dimensional calculation.

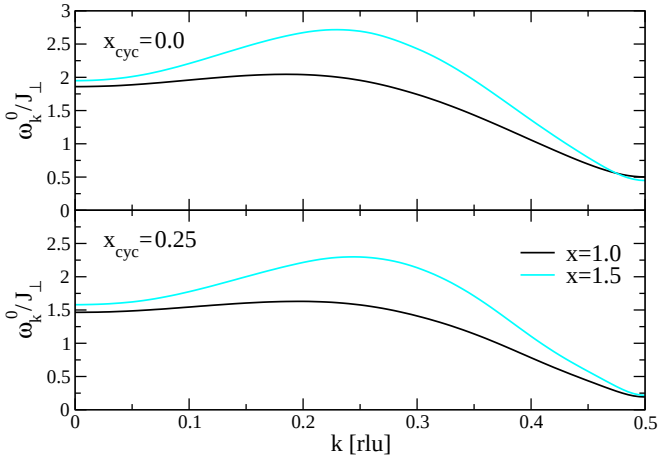

FIG. 3: Dispersion ω_k^0 of a single triplon for an isolated ladder. *Upper panel:* One-triplon dispersion for $x_{\rm cyc} = 0$ and $x = 1$ (black curve) and $x = 1.5$ (cyan/grey curve). *Lower panel:* One-triplon dispersion for $x_{\rm cyc} = 0.25$ and $x = 1$ (black curve) and $x = 1.5$ (cyan/grey curve).

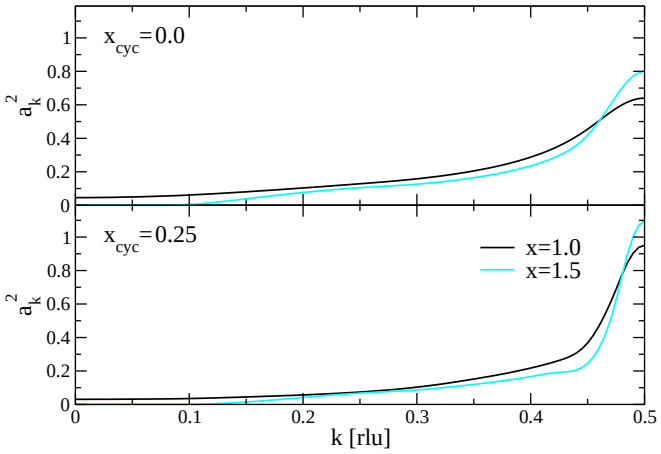

FIG. 4: Weight a_k^2 of a single triplon for an isolated ladder. *Upper panel:* One-triplon spectral weight for $x_{\rm cyc} = 0$ and $x = 1$ (black curve) and $x = 1.5$ (cyan/grey curve). *Lower panel:* One-triplon spectral weight for $x_{\rm cyc} = 0.25$ and $x = 1$ (black curve) and $x = 1.5$ (cyan/grey curve).

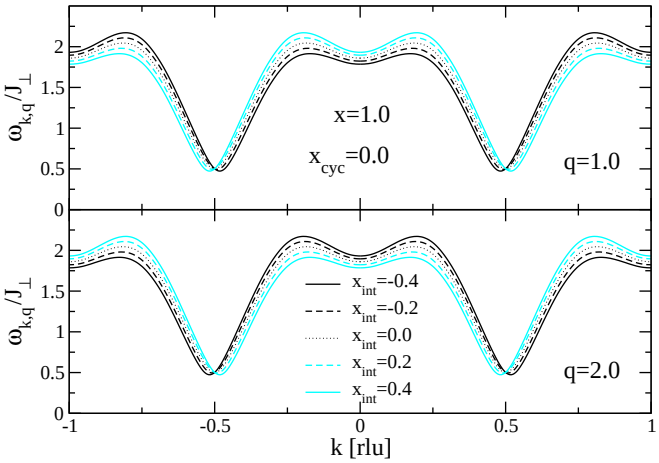

FIG. 5: The influence of the inter-ladder coupling $x_{\rm int}$ on the dispersion $\omega_{k,q}$ of a single triplon on the trellis lattice as shown in Figs. 2 for $x = 1$ and $x_{\rm cyc} = 0$. *Upper panel:* Data sets for $q = 1$ and $x_{\rm int} = \{-0.4; -0.2; 0.0; 0.2; 0.4\}$. *Lower panel:* Data sets for $q = 2$ and $x_{\rm int} = \{-0.4; -0.2; 0.0; 0.2; 0.4\}$.

Note that at $x = 1.5$ the extrapolation of the weight at small momentum spuriously leads to slightly negative values. In this parameter regime the three-triplon continuum overlaps with the one-triplon dispersion leading to a possible decay and a finite life-time of the elementary excitations. Therefore we put the one-triplon spectral weight in this range to zero.[23] We refrain from displaying data for even larger values of x because there is a growing body of evidence that $x \in (1, 1.5)$ is the relevant range for cuprate ladders[4,5,6,7,19,20] even though early quantum chemical calculations[21] indicated even larger values of $x \approx 2$. Moreover, our approach is less reliable beyond $x \approx 1.5$.

In the following, we will first discuss the generic properties of the two-dimensional spectra. To this end, we concentrate on the case $x = 1$ and $x_{\rm cyc} = 0$. But the features obtained are similar for all couplings in the paramagnetic phase. The influence of the inter-ladder coupling $x_{\rm int}$ on the dispersion of a single triplon is shown in Fig. 5. For $q = 1.5$ the effect of the trellis structure vanishes completely due to destructive interference. The largest effects can be seen for $q = 1$ and $q = 2$. (We refer to $q = 1.5$ and $q = 2$ instead of $q = 0.5$ and $q = 0$ although this does not make any difference in the framework of our theoretical model. But it eases the contact to the experimental investigations, cf. e.g. Ref. 7, where one keeps away from zero momentum where no magnetic scattering occurs.) Clearly, deviations from $q = 1.5$ lead to a lifting of the reflection symmetry about $k = \pm 0.5$ and to a difference between $q = 1$ and $q = 2$. On the level of our description, a ferromag-

netic coupling $-|x_{\rm int}|$ at $q=1$ is equivalent to an antiferromagnetic coupling $|x_{\rm int}|$ at $q=2$ and vice versa. The one-triplon gap is shifted for an antiferromagnetic coupling from $k=0.5$ to a *lower* momentum for $q=2$ and from $k=0.5$ to a *higher* momentum for $q=1$.

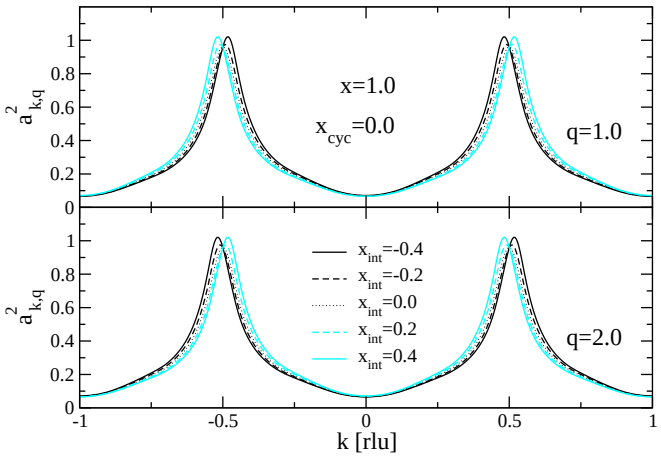

FIG. 6: The influence of the inter-ladder coupling $x_{\rm int}$ on the spectral weight $a^2_{k,q}$ of a single triplon on the trellis lattice as shown in Figs. 2 for $x=1$ and $x_{\rm cyc}=0$. *Upper panel:* Data sets for $q=1$ and $x_{\rm int}=\{-0.4;-0.2;0.0;0.2;0.4\}$. *Lower panel:* Data sets for $q=2$ and $x_{\rm int}=\{-0.4;-0.2;0.0;0.2;0.4\}$.

The physical interpretation of the destructive interference at half-integer values of q or of k relies on the geometry of the trellis lattice, see Fig. 2. Consider, for instance, two hopping processes from the two rungs, where the arrows e_1 and e_2 end, to the rung, from where both arrows start. For half-integer value of q these processes have opposite sign so that they cancel each other. Hence no effect of $J_{\rm int}$ occurs. The same is true for two hopping processes from the two rungs in the ladder in the middle in Fig. 2 to one of the other rungs. For half-integer value of k these processes have opposite sign so that they cancel each other. Again, no effect of $J_{\rm int}$ occurs. This explains why $J_{\rm int}$ has to be significantly large in order to imply a sizeable effect if at least one of the momenta q and k is close to a half-integer values. Additionally, Fig. 4 shows that for values of k which deviate substantially from half-integer values the spectral weight of the triplons is very low so that the inter-ladder coupling has only very limited influence on their dynamics, even away from half-integer values of momentum k.

Besides the energy of the elementary triplon excitation, also the spectral weight is affected by the inter-ladder coupling. The correspoding results are shown in Fig. 6. In accordance to the one-triplon gap, the maximum of the one-triplon spectral weight is shifted for an antiferromagnetic coupling from $k=0.5$ to a lower momentum k for $q=2$ and from $k=0.5$ to a higher momentum k for $q=1$. But the overall effect on this quantity is rather small due to the frustration.

Next we discuss a possible experimental detection of the inter-ladder coupling in inelastic neutron scattering experiments. From the above, it becomes clear that measurements collecting events in the interval $q\in[1,2]$ do not allow to establish the inter-ladder coupling. We consider the possible detection of an inter-ladder coupling to be an essential question because it concerns the dimensionality of the magnetic systems $A_{14}Cu_{24}O_{41}$ and $SrCu_2O_3$. Therefore, we suggest to measure around $q=1$ and around $q=2$. The comparison of the two constant energy scans of k provide the data to determine the value of $J_{\rm int}$. We point out that the weight factor $\sin^2\left(\pi q\frac{a}{2a_2}\right)$ is reduced only by 25% if one deviates from the optimum value of $q=1.5$ (where the sine is unity) to $q=1$ or $q=2$. In this argument, we assume the approximate ratio $a_2=1.5a$.

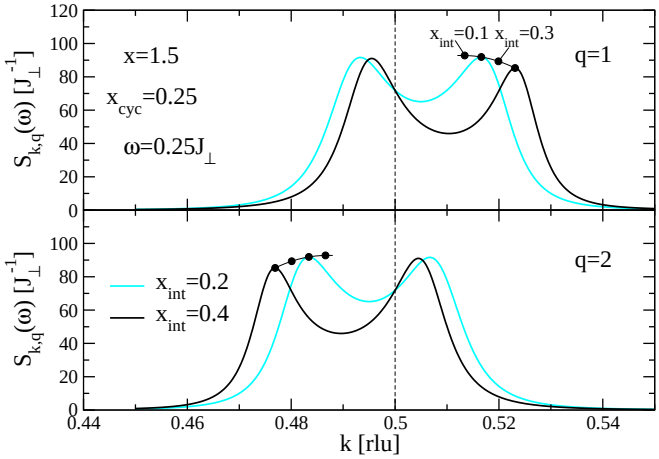

FIG. 7: Constant ω-scan of the dynamical structure factor at $\omega=0.25J_\perp$ with $x=1.5$ and $x_{\rm cyc}=0.25$ as a function of the momentum k in ladder direction. The black curves correspond to $x_{\rm int}=0.4$ and the cyan/grey curves correspond to $x_{\rm int}=0.2$. The uppper panel shows $q=1$ and the lower panel shows $q=2$. A Lorentzian broadening of $\eta=0.05$ is used in all panels.

In our opinion, the best experimental way to determine the inter-ladder coupling by inelastic neutron scattering is to perform constant energy scans $S(k,q=\pm1,\omega=\Delta+\delta)$ at momenta $q=1$ and $q=2$ for energies $\Delta+\delta$ slightly above the one-triplon gap Δ, i.e. $\delta>0$. The resulting spectra will show characteristic asymmetries with respect to reflections about $k=0.5$, see Fig. 7.

The existing experimental data of magnetic excitations in $A_{14}Cu_{24}O_{41}$ and $SrCu_2O_3$ are consistent with coupling constants $x=1.2-1.5$ and $x_{\rm cyc}=0.2-0.25$[5,6,7]. In the following, we discuss the parameter set $x=1.5$ and $x_{\rm cyc}=0.25$ which has been deduced from recent inelastic neutron scattering data for $La_4Sr_{10}Cu_{24}O_{41}$[7]. Typical curves are shown in Fig. 7. Note again that in our mean-field treatment a change from antiferromagnetic to ferromagnetic inter-ladder coupling $|x_{\rm int}|\to-|x_{\rm int}|$ leads to the same curves as in Fig. 7 where the momentum q is changed to $q+1$.

The large anisotropy $x=J_\parallel/J_\perp=1.5$ is questioned by quantum chemistry calculations[8]. It is argued that the magnetic exchanges along the legs $J_\parallel$ and the rungs $J_\perp$ are almost equal but a strong ferromagnetic inter-ladder coupling $x_{\rm int}\approx-0.2$ is present (the calculations were done for the compound $SrCu_2O_3$). We want to point out that this scenario of an

isotropic exchange $x \approx 1$ is *not* consistent with our results. As discussed above, the effects of the inter-ladder coupling are washed out if one averages over the momentum q perpendicular to the ladder direction. This has been done when fitting the experimental data in Ref. 7. Therefore, the deduced parameters $x = 1.5$ and $x_{\rm cyc} = 0.25$ will not change if one takes into account a finite coupling $x_{\rm int}$.

Independently from the intra-ladder anisotropy x, the strong ferromagnetic inter-ladder coupling deduced in Ref. 8 is an interesting new aspect of the physics in these materials. It will be interesting to see if the analysis of inelastic neutron scattering data using the results obtained in this work will yield values of the magnetic exchange $J_{\rm int}$ which are consistent with the quantum chemistry calculations.

In summary, we have discussed the properties of magnetic excitations on the frustrated trellis lattice in the paramagnetic phase by calculating the one-triplon dispersion and the corresponding one-triplon contribution to the dynamical structure factor. The trellis lattice can be viewed as being built from coupling components of one-dimensional two-leg ladders. Technically, an effective model for a single two-leg ladder is used as the starting point to discuss the influence of the inter-ladder coupling. The strong frustration of the lattice causes the system to be effectively one-dimensional, i.e. the magnetic excitation spectrum reveals many properties of a single two-leg spin ladder. The leading order of the inter-ladder coupling $J_{\rm int}$ interferes destructively around the minima of the dispersion in the ladder. Hence, the effects of small or even sizable inter-ladder couplings are generically small. Yet, they lead to typical asymmetries of the dispersion and the spectral weight of a single mode.

Although the mobility of triplons is strongly reduced by frustration, we expect that two- or multi-triplon properties like the *interaction* between the triplons are affected in a stronger fashion. While the hopping of triplons cancels due to frustration their interaction as induced by different couplings adds. Thus, it is an important challenge for future work to clarify the influence of the inter-ladder coupling on optical properties and on all other probes of multi-triplon properties.

We have made a detailed prediction how to detect the magnetic inter-ladder coupling in experimental systems like the so-called telephone-number compounds $A_{14}Cu_{24}O_{41}$ and the system $SrCu_2O_3$. We have provided arguments that experimental averages over the momentum perpendicular to the ladder direction do not yield information about the inter-ladder coupling. Therefore, the recently deduced parameter set $x = 1.5$ and $x_{\rm cyc} = 0.25$ will not be affected by a finite inter-ladder coupling[7]. This experimentally established difference between $J_\parallel$ and $J_\perp$ calls for further quantum chemical analyses of the superexchange couplings.

The study of the inelastic neutron scattering spectra at $q = 1$ and $q = 2$ should clarify the sign and the size of the two-dimensional inter-ladder coupling in the experimental systems. Thereby, the imporant issue of the dimensionality of these magnetic systems will be elucidated.

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22 The skewed unit cell as shown in Fig. 2 is simpler in spite of the 'unusual' periodicities because it contains only one dimer so that it is a Bravais lattice with one triplon mode per unit cell. If one insisted on using unit vectors along and perpendicular to the ladders the unit cell would comprise two dimers so that two modes per unit cell have to be considered. They correspond to the

modes at q and at $q+1$.

[23] Recall that the overall total spectral weight at small momentum is small anyway. Additionally, the three-triplon contribution to the dynamical structure factor behaves like ω^3 close to the lower band edge leading to a long life-time and therefore a sharp resonance at small momentum. This high power results from the hardcore property of the triplons.